FORM PTO-1390 (REV. 6-87)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

1022-01

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US)

71/31/99/04214	NTERNATIONAL APPLICATION NO.		L FILING DATE	PRIORITY DAT	TE CLAIN	MED	
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8. ☐ A translation of the Annexes to 36 (35 U.S.C. 371(c)(5)).	the International Preliminary Examination Report under PCT Article
Other document(s) or information inc	uded:
9. □ An Information Disclosure Stat	ement under 37 C.F.R. 1.97 and 1.98.
L1. The above checked items are being a. □ before the 18th month publication and the Articolor. □ after 20 months but before 22 d. □ after 22 months (surcharge and Note: Petition to revive (37 C. months and no proper demand claimed priority date. e. ■ by 30 months and a proper demand claimed priority date. e. ■ by 30 months and a proper demand claimed priority date. g. □ after 30 months but before 32 by the 19th month from the earliest claimed priority date.	tion. the 20 communication but before 20 months from the priority date. months (surcharge and/or processing fee included). d/or processing fee included). F.R. 1.137(a) or (b)) is necessary if 35 U.S.C. 371 requirements submitted after 22 for International Preliminary Examination was made by 19 months from the earliest mand for International Preliminary Examination was made by the 19th month from the. months and a proper demand for International Preliminary Examination was made diest claimed priority date (surcharge and/or processing fee included)
 At the time of transmittal, the time a. □ has expired and no amendmen b. □ has not yet expired. 	limit for amending claims under Article 19 ts were made.
3. Certain requirements under 35 l	J.S.C. 371 were previously submitted by the applicant on, namely:
	SCHNADER HARRISON SEGAL & LEWIS
Pate:	Austin R. Miller, Reg. No. 16,602 1600 Market Street, 36th Floor

Philadelphia, PA 19103

DESCRIPTION

OPEN-ENDED POLYIMIDE MOLDINGS AND METHOD FOR PRODUCING THEM

TECHNICAL FIELD

The present invention relates to open-ended polyimide moldings with good heat resistance and electric insulation properties.

PRIOR ART

With good heat resistance, polyimides have many applications for heat insulation, for example, for heat-insulating films for parts of electronic appliances, for heat-resistant parts of electric appliances, etc. At present, however, polyimides could not be formed into thin-walled moldings or large-sized moldings on an industrial scale, as their workability are poor.

One conventional method for producing relatively thin-walled polyimide moldings comprises molding aromatic polyimide powder through compression or sintering at high temperatures not lower than 450°C. Another method is known, which comprises molding polyimides relatively easy to soften, such as bismaleimides or polyethers, through compression or injection. Needless-to-say, these methods are for in-mold working.

Still another method is known, comprising applying a polyimide precursor, polyamic acid onto the surface of a desired mold to form a film thereon, followed by curing it under heat to give a polyimide molding.

On the other hand, further known is a method comprising molding a polyimide film having been prepared by sheeting a polyamic acid on a metal support, for which is used a female-male mold under heat and pressure.

The open-ended polyimide moldings thus produced include speaker diaphragms, reflectors for lighting appliances, piezoelectric devices for surface mounting, etc.

The prior art techniques noted above are effective in their own ways for the intended applications.

According to the prior art techniques, however, moldings, especially those having a wall thickness of not larger than 0.5 mm are difficult to produce, and it has heretofore been said that producing such thin-walled moldings on an industrial scale is impossible. The problems with the method of curing polyimide precursor films on molds are that uniform films could not be formed and that the films formed often have defects such as pin holes, etc. According to the method, therefore, complicated moldings with grooved or hilled surface profiles could not be obtained. The method of producing polyimide films through molding under heat or pressure requires heating the entire mold used therein. In

the method, therefore, moldings with large opening area or large surface area are difficult to be produced. As a rule, polyimide films are not thermoplastic, and could not elongate sufficiently when softened under heat. Therefore, producing deep-drawn moldings of polyimide films is impossible.

Accordingly, the object of the invention is to solve the defects in the prior art noted above and to provide thin-walled, preferably deep-drawn, open-ended polyimide moldings and a method for producing them.

DISCLOSURE OF THE INVENTION

To attain the object noted above, the invention includes the following means:

- (1) An open-ended polyimide molding of an aromatic polyimide resin, which is characterized in that its wall thickness is at most 0.5 mm, and that the ratio of its depth to its opening is at least 0.7, or its longest major axis is at least 150 mm in length with its draw depth being at least 0.5 mm.
- (2) The open-ended polyimide molding of above (1), which is such that its wall thickness falls between 0.001 and 0.3 mm, and that the ratio of its depth to its opening falls between 0.7 and 5.0, or its longest major axis falls between 150 and 10000 mm in length with its draw depth falling between 0.5 and 8000 mm.

- (3) The open-ended polyimide molding of above (1), which is such that its wall thickness falls between 0.01 and 0.2 mm, and that the ratio of its depth to its opening falls between 1.0 and 3.0, or its longest major axis falls between 200 and 5000 mm in length with its draw depth falling between 1.0 and 2000 mm.
- (4) The open-ended polyimide molding of any one of above (1) to (3), of which the aromatic polyimide is a thermoplastic aromatic polyimide.
- (5) The open-ended polyimide molding of above (4), of which the thermoplastic polyimide has a glass transition temperature falling between 200 and 350°C and has a degree of elongation at break of from 50 to 2000 % at its glass transition temperature.
- (6) A method for producing an open-ended polyimide molding having a wall thickness of at most 0.5 mm, which is characterized by forming a thermoplastic polyimide film in vacuum into its molding.
- (7) The method for producing an open-ended polyimide molding of above (6), in which the molding produced is such that the ratio of its depth to its opening is at least 0.7, or its longest major axis is at least 150 mm in length with its draw depth being at least 0.5 mm.
- (8) The method for producing an open-ended polyimide molding of above (6) or (7), in which the molding produced

includes a plurality of repetitive patterns.

(9) The method for producing an open-ended polyimide molding of above (8), in which the molding is produced in one vacuum forming operation.

BEST MODES OF CARRYING OUT THE INVENTION

The invention is described concretely hereinunder.

The aromatic polyimide is a condensate of an aromatic tetracarboxylic acid and an aliphatic or aromatic diamine. Typically, it is obtained by polycondensing a tetracarboxylic acid dianhydride such as pyromellitic acid dianhydride, biphenyltetracarboxylic acid dianhydride or the like with a diamine such as paraphenylenediamine, diaminodiphenyl ether or the like to give a polyamic acid, followed by curing it for ring closure under heat or with a catalyst. In the invention, preferred are thermoplastic aromatic polyimides. To obtain thermoplastic polyimides, for example, the following compounds may be copolymerized.

Dicarboxylic acid anhydrides for that purpose include pyromellitic acid dianhydride, 4,4'-hydroxydiphthalic acid dianhydride, 3,3',4,4'-benzophenonetetracarboxylic acid dianhydride, 2,2',3,3'-benzophenonetetracarboxylic acid dianhydride, 3,3',4,4'-biphenyltetracarboxylic acid dianhydride, 2,2',3,3'-biphenyltetracarboxylic acid dianhydride, 2,2',3,3'-biphenyltetracarboxylic acid dianhydride, 2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane

dianhydride, bis(3,4-dicarboxyphenyl) sulfone dianhydride, bis(3,4-dicarboxyphenyl) sulfide dianhydride, bis(2,3dicarboxyphenyl)methane dianhydride, bis(3,4dicarboxyphenyl)methane dianhydride, 1,1-bis(2,3dicarboxyphenyl)methane dianhydride, 1,1-bis(2,3dianhydride, dicarboxyphenyl)propane 2,2-bis(3,4dicarboxyphenyl)propane dianhydride, m phenylenebis(trimellitic acid) dianhydride, etc.

include Diamines hexamethylenediamine, heptamethylenediamine, 3,3'-dimethylpentamethylenediamine, 3-methylhexamethylenediamine, 3 methylheptamethylenediamine, 2,5dimethylhexamethylenediamine, octamethylenediamine, nonamethylenediamine, 1,1,6,6tetramethylhexamethylenediamine, 2,2,5,5tetramethylhexamethylenediamine, 4,4dimethylheptamethylenediamine, decamethylenediamine, phenylenediamine, 4,4'-diaminobenzophenone, 4-aminophenyl 3-aminobenzoate, m-aminobenzoyl-p-aminoanilide, diaminodiphenyl ether, 3,4'-diaminodiphenyl ether, bis(4aminophenyl)methane, 1,1-bis(4-aminophenyl)ethane, bis(4-aminophenyl)propane, 4,4'-diaminodiphenyl sulfoxide, 3,3'-diaminobenzophenone, 1,3-bis(4-aminophenoxy)benzene, 2,2'-diaminobenzophenone, 1,2-bis(4-aminophenoxy)benzene, 1,3-bis(4-aminobenzoyloxy)benzene, 4,4'-dimainobenzanilide, 4,4'-bis(4-aminophenoxy)phenyl ether, 2,2'-bis(4-aminophenyl)hexafluoropropane, 2,2'-bis(4-aminophenyl)1,3-dichloro-1,1,3,3-tetrafluoropropane, 4,4'diaminodiphenyl sulfone, 1,12-diaminododecane, 1,13diaminododecane, polysiloxanediamine, etc.

Of the compounds noted above, preferred for use in the invention are copolymers of 1,3-bis(4-aminophenoxy)benzene (referred to as RODA), pyromellitic acid dianhydride (referred to as PMDA) and 4,4'-hydroxydiphthalic acid dianhydride; polymers of 4,4'-diaminodiphenyl ether (referred to as ODA) and 3,3',4,4'-biphenyltetracarboxylic acid dianhydride (referred to as BPDA); and copolymers of ODA, PMDA and BPDA.

Thermoplastic aromatic polyimides soften when heated. Preferred for use in the invention are those having a glass transition point falling between 200 and 350°C, more preferably between 220 and 300°C. Also preferred are those having a degree of elongation at break at their glass transition temperature of from 50 to 2000 %, more preferably from 300 to 800 %.

The invention is to obtain open-ended moldings with no closed part, typically including tray-shaped, carrier belt-shaped or cup container-shaped moldings to be produced by deforming or drawing films. Needless-to-say, the open-ended moldings may be hot-sealed and integrated into closed moldings, owing to the hot-sealability of thermoplastic polyimides. The invention shall encompass such closed

moldings. The method for obtaining the moldings is not specifically defined, to which is applicable any of vacuum forming, injection molding and the like or even their combination to give polyimide film-coated injection moldings.

The open-ended polyimide moldings of the invention have a wall thickness of at most 0.5 mm, preferably from 0.001 to 0.3 mm, more preferably from 0.01 to 0.2 mm.

The open-ended polyimide moldings of the invention are such that the ratio of the depth to the opening thereof is at least 0.7, or the longest major axis thereof is at least 150 mm in length with the draw depth thereof being at least 0.5 mm. For the moldings with the ratio of the depth to the opening thereof being at least 0.7, the length of the opening is not defined, but preferably, the draw depth ratio falls between 0.7 and 5.0, more preferably between 1.0 and 3.0. For the moldings with the longest major axis thereof being at least 150 mm in length, it is not always necessary that the ratio of the depth to the opening thereof is at least 0.7, but the draw depth thereof shall be at least 0.5 mm, preferably falling between 0.5 and 8000 mm, more preferably between 1.0 and 2000 mm.

In short, the invention is directed to thin-walled, open-ended polyimide moldings of two types both having a wall thickness of at most 0.5 mm, but one type being deep-drawn moldings having a draw ratio of at least 0.7, and the other

being non-deep-drawn but large-sized moldings having a longest major axis of at least 150 mm in length.

The method for producing the open-ended polyimide moldings of the invention comprises forming a thermoplastic polyimide film in vacuum into its molding having a wall thickness of at most 0.5 mm. The open-ended polyimide moldings produced in the method may well have a wall thickness of at most 0.5 mm, but are preferably deep-drawn moldings having a draw ratio of at least 0.7, or non-deep-drawn but large-sized moldings having a longest major axis of at least 150 mm in length.

Vacuum forming includes a straight method, a draping method, an air-slip method, a snap-back method, a plug-assisted method, etc., any of which could apply to the invention. Apart from those, a pressure forming method could also apply to the invention. Accordingly, the invention encompasses the mode of pressure forming as one type of vacuum forming. The moldings produced according to the method of the invention may include a plurality of repetitive patterns. A series of plural moldings each having a predetermined form profile may be produced in the method, and it may be separated into individual moldings. In this case, the longest major axis of the series of plural moldings produced shall be at least 150 mm in length. In this case, however, patterning the series of plural moldings is preferably finished in one vacuum forming operation. This

means that the method of that case is not for successively forming a pattern of each molding one after another on a film. However, for a carrier belt for which the polyimide molding shall have repetitive patterns and shall be long and windable into a roll, the total length of the carrier belt shall differ from the length of the longest major axis defined herein. Namely, the longest major axis referred to in the invention for the profile of such a carrier belt indicates the length of one molding that includes a plurality of repetitive patterns to be formed in one molding operation. Therefore, the carrier belt itself could be obtained by repeating a plurality of times the forming operation of the invention (that is, the operation of forming a plurality of patterns) on a long film.

EXAMPLE

The invention is described in more detail with reference to the following Example. In the Example, the glass transition temperature of each sample was measured through DSC, and the degree of elongation thereof at break was measured according to the method mentioned below.

Degree of Elongation at Break:

A thermostat in which the temperature difference of 5°C can be controlled is first heated up to the glass transition temperature (Tg) of the sample to be tested, and a part of a tensile tester (defined in JIS C-2381) with the sample mounted

thereon is inserted into the thermostat. In that condition, after the film has reached its Tg (after about 1 hour), the degree of elongation of the sample is measured according to JIS C-2318.

Example 1:

A thermoplastic aromatic polyimide film of 0.075 mm thick ("Kapton" 300KJ from DuPont, having a glass transition temperature of 220°C and having a degree of elongation at break at 220°C of 550 %) was fixed to a metallic frame with its periphery being held by the frame, and the center part of the film was heated at 280°C. In that condition, the film was grounded on a female mold equipped with a degassing mechanism, and the mold was degassed to attain vacuum forming of the film. One mold used herein had an opening of 200 mm and a draw depth of 70 mm; and the other had an opening of 35 mm and a draw depth of 52.5 mm. The both were patterned open-type molds, and the latter was a drawing mold having a ratio of the depth to the opening of 1.5. The moldings thus produced through vacuum forming were almost uniform in thickness, and correctly received the mold pattern.

INDUSTRIAL APPLICABILITY

The invention provides thin-walled, deep-drawn or large-sized, open-ended polyimide moldings which have good heat resistance and electric insulation properties and which

are easy to produce through vacuum forming. The moldings are usable, for example, for speaker diaphragms, reflectors for lighting appliances, piezoelectric devices for surface mounting, etc.

CLAIMS

- 1. An open-ended polyimide molding of an aromatic polyimide resin, which is characterized in that its wall thickness is at most 0.5 mm, and that the ratio of its depth to its opening is at least 0.7, or its longest major axis is at least 150 mm in length with its draw depth being at least 0.5 mm.
- 2. The open-ended polyimide molding as claimed in claim 1, which is such that its wall thickness falls between 0.001 and 0.3 mm, and that the ratio of its depth to its opening falls between 0.7 and 5.0, or its longest major axis falls between 150 and 10000 mm in length with its draw depth falling between 0.2 and 8000 mm.
- 3. The open-ended polyimide molding as claimed in claim 1, which is such that its wall thickness falls between 0.01 and 0.2 mm, and that the ratio of its depth to its opening falls between 1.0 and 3.0, or its longest major axis falls between 200 and 5000 mm in length with its draw depth falling between 1.0 and 2000 mm.
- 4. The open-ended polyimide molding of any one of claims 1 to 3, of which the aromatic polyimide is a thermoplastic aromatic polyimide.
- 5. The open-ended polyimide molding as claimed in claim 4, of which the thermoplastic polyimide has a glass transition temperature falling between 200 and 350°C and has a degree of

elongation at break of from 50 to 2000 % at its glass transition temperature.

- 6. A method for producing an open-ended polyimide molding having a wall thickness of at most 0.5 mm, which is characterized by forming a thermoplastic polyimide film in vacuum into its molding.
- 7. The method for producing an open-ended polyimide molding as claimed in claim 6, in which the molding produced is such that the ratio of its depth to its opening is at least 0.7, or its longest major axis is at least 150 mm in length with its draw depth being at least 0.5 mm.
- 8. The method for producing an open-ended polyimide molding as claimed in claim 6 or 7, in which the molding produced includes a plurality of repetitive patterns.
- 9. The method for producing an open-ended polyimide molding as claimed in claim 8, in which the molding is produced in one vacuum forming operation.

ABSTRACT

An open-ended polyimide molding of an aromatic polyimide resin, which is characterized in that its wall thickness is at most 0.5 mm, and that the ratio of its depth to its opening is at least 0.7, or its longest major axis is at least 150 mm in length with its draw depth being at least 0.5 mm.

	Attorney Docket No
	Original Application
	PCT National Application
	U.S. Designated Office Continuation or Divisional Application
	Continuation of Divisional Application Continuation-in-Part Application
-	COMBINED DECLARATION, POWER OF ATTORNEY AND PETITION
As a belo	ow named inventor, I hereby declare that:
My resid	ence, post office address and citizenship are as stated below next to my name,
(if plural	I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor l names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention OPEN-ENDED POLYIMIDE MOLDINGS AND METHOD FOR PRODUCING THEM
☐ whice	ch is described in the specification and claims
	☐ attached hereto.
	☐ filed on
	Application Serial No.
	and was amended on
	(if applicable)
■ whi	ich is described in International Application No. PCT/JP99/04214
filed _	August 4, 1998 and as amended on
1	(if any),
which I	have reviewed and for which I solicit a United States patent.

1022-01

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which I have reviewed and for which I solicit a United States patent.

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I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe that this invention was ever known or used in the United States before my or our invention thereof or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application or said international application, or in public use or on sale in the United States of America more than one year prior to this application or said international application, or that the invention has been patented or made the subject of an inventor's certificate issued before the date of this application or said international application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months prior to this application or said international application, or that any application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application or said international application by me or my legal representatives or assigns except as identified below.

COMBINED DECLARATION, POWER OF ATTORNEY AND PETITION (Page 2)

Attorney	Docket No.		

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International Application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application(s) for patent or inventor's certificate, or of any PCT International Application having a filing date before that of the application on which priority is claimed:

Number	Country	Date of Filing (day,month,year)	Priority Claimed
H10-233628	Japan	4, August, 1998	■ yes □ no
			□ yes □ no
			□ yes □ no
			□ yes □ no
			□ yes □ no
(Application Serial No.)	(Filing L	Date) (Si	tatus)(patented,pending,abandone
Application Serial No.) Application Serial No.)	(Filing L		tatus)(patented,pending,abandone tatus)(patented,pending,abandone
Application Serial No.) POWER OF ATTORNE	(Filing L Y: As a named inventor, I he ared attorneys to prosecute the		tatus)(patented,pending,abandone
POWER OF ATTORNE and the following register frademark Office connects. Daniel Christenbury Guy T. Donatiello Paul A. Taufer James A. Drobile Austin R. Miller Gerard J. Weiser	(Filing L. Y: As a named inventor, I have attorneys to prosecute the sted therewith: Reg. No. 31,750 Reg. No. 33,167 Reg. No. 35,703 Reg. No. 19,690 Reg. No. 16,602 Reg. No. 19,763	Patrick J. Farley Reg. No. Michael A. Patane Reg. No. David A. Sasso Reg. No. Robert A. McKinley Reg. No. Sharon Fenick Reg. No.	tatus)(patented,pending,abandone
POWER OF ATTORNE and the following register frademark Office connects. Daniel Christenbury Guy T. Donatiello Paul A. Taufer James A. Drobile Austin R. Miller	(Filing L. Y: As a named inventor, I have attorneys to prosecute the sted therewith: Reg. No. 31,750 Reg. No. 33,167 Reg. No. 35,703 Reg. No. 19,690 Reg. No. 16,602 Reg. No. 19,763 Reg. No. 38,940	Patrick J. Farley Reg. No. Michael A. Patane Reg. No. David A. Sasso Reg. No. Robert A. McKinley Reg. No. Sharon Fenick Reg. No.	eys listed under Customer No. 022 siness in the United States Patent o. 42,524 o. 42,982 o. 43,084 o. 43,793 o. 45,269 o. 46,201

COMBINED DECLARATION, POWER OF ATTORNEY AND PETITION (Page 3)

Attorney Docket No. 1022-01

I hereby petition for grant of a United States Letters Patent on this invention.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

thereon.		
1. FULL NAME OF SOLE OR FIRST INVENTOR	INVENTOR'S SIGNATURE DATE	: 2001
Hideaki Machida		22th, ,2001
RESIDENCE	CITIZENSHIP	
Yokohama, Japan	Japanese	
POST OFFICE ADDRESS	nagawa 234-0052 Japan	
,		
2. FULL NAME OF JOINT INVENTOR, IF ANY Hirokazu Yokoyama	INVENTOR'S SIGNATURE, DATE	/i ≥ /2001
	CITIZENSHIP	1161
RESIDENCE Handa, Japan	Japanese	
POST OFFICE ADDRESS 100-2, Yanabe Takayamacho 3-chome, Handa-shi, Aicl	ni 475-0062 Japan	
···	INVENTOR'S SIGNATURE	DATE
3. FULL NAME OF ADDITIONAL JOINT INVENTOR, IF ANY	IMAEM TOK 2 SIGNAT OKE	DAIE
RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS		
4. FULL NAME OF ADDITIONAL JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS	1	
5. FULL NAME OF ADDITIONAL JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE	CITIZENSHIP	•
POST OFFICE ADDRESS		
6. FULL NAME OF ADDITIONAL JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS		
7. FULL NAME OF ADDITIONAL JOINT INVENTOR, IF ANY	INVENTOR'S SIGNATURE	DATE
RESIDENCE	CITIZENSHIP	
POST OFFICE ADDRESS		

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